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Superconducting state in (W, Ta)$_5$SiB$_2$

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Abstract

We characterize the superconducting state in a boro-silicide (W, Ta)$_5$SiB$_2$, with T_c of 6.5 K by means of magnetization, electrical resistivity, and specific heat measurements. As x increased, the transition temperature T_c abruptly enhances from 5.8 to 6.5 K. The magnetization versus magnetic field (M-H) curve indicated that (W, Ta)$_5$SiB$_2$ was a conventional type-II superconductor. The estimated lower critical field $H_{c1}(0)$ and upper critical field $H_{c2}(T)$ are about 121 Oe and 14.7 kOe, respectively. The penetration depth $\lambda(0)$ and coherence length $\xi(0)$ are calculated to be approximately 369 and 14.9 nm, respectively, using Ginzburg-Landau (GL) equations. Specific heat data shows the superconductivity in W$_{4.5}$Ta$_{0.5}$SiB$_2$ belongs to a week-coupling BCS superconductor. Finally, we discuss the increasing of T_c in of (W, Ta)$_5$SiB$_2$ system.

Keywords: superconductivity; T$_2$ phase; W$_{5-x}$Ta$_x$SiB$_2$

1. Introduction

Superconductor of layered transition metal boro-silicide TM$_5$SiB$_2$ (TM = Nb, Mo, and W) which is called as T$_2$ phase has been recently discovered [1,2,3]. TM$_5$SiB$_2$ is crystallized in a tetragonal structure (Mo$_5$SiB$_2$-type) with space group: *I*4/*mcm* (No. 140). Superconducting transition temperature (T_c) of T$_2$ phase superconductors Nb$_5$SiB$_2$, Mo$_5$SiB$_2$, and W$_5$SiB$_2$ are 7 K, 5.6 K, and 5.8 K respectively. In TM = 4d transition metal elements of Nb and Mo system, T_c of Nb$_5$SiB$_2$ is larger than that of Mo$_5$SiB$_2$. Moreover, in TM = W system, T_c value increases from 5.8 K to 6.5 K by partially substituting Ta for W [4]. However, there are few information of superconducting state in W$_{5-x}$Ta$_x$SiB$_2$ about superconducting parameter and increasing of T_c in W$_5$SiB$_2$. In this paper, we successfully synthesized the polycrystalline sample of sample of (W, Ta)$_5$SiB$_2$ with T_c = 6.5 K and performed the magnetic and transport properties measurements in superconducting state to determine the superconducting parameters. Moreover, we discuss the origin of increasing of T_c comparison the specific heat results of W$_5$SiB$_2$ and W$_{4.5}$Ta$_{0.5}$SiB$_2$.

2. Experimental details

Polycrystalline sample of W$_{5-x}$Ta$_x$SiB$_2$ was synthesized by the conventional solid state reaction. We confirmed that synthesized sample included W$_{5-x}$Ta$_x$SiB$_2$ as main phase and WSi$_2$ as minor impurity phase. The impurity phase contained in this sample is ~20%. The sample quality of W$_{5-x}$Ta$_x$SiB$_2$ is similar in purity of non-substituted sample

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W_5SiB_2 [3]. The magnetizations were determined by SQUID magnetometer (MPMS-R2, Quantum Design) at temperatures between 1.8 K and 10 K in magnetic fields up to 15 kOe. The electrical resistivity was measured using a conventional dc four-probe method with the measuring current $I = 1.0$ mA in the temperature range from 0.8 to 20 K at applied magnetic fields up to 20 kOe, using PPMS (Quantum Design) and a laboratory-built apparatus. Electrical leads were fabricated by spot-welding gold wires (25μm) onto the polished surface of a specimen. The specific heat measurements were performed using PPMS (Quantum Design) in temperature range of 0.5-10 K under zero and an applied magnetic field.

3. Experimental results and discussion

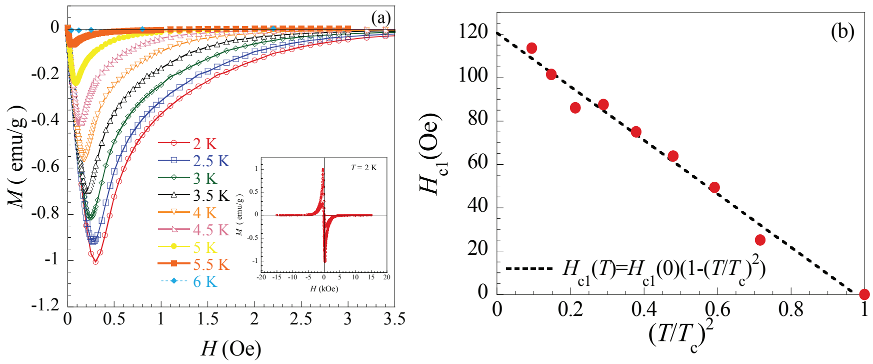

Fig. 1. (Color online) (a) Magnetization vs magnetic field curves of $W_{4.5}Ta_{0.5}SiB_2$ at various temperatures. Inset shows the *M-H* curve at high magnetic field range. (b) $H_{c1}(T)$ as a function of $(T/T_c)^2$.

Figure 1 shows the magnetic field dependence of the magnetization of $W_{4.5}Ta_{0.5}SiB_2$ (nominal composition) measured at various temperatures. The data in the inset of Fig. 1 shows a hysteresis, indicating that $W_{4.5}Ta_{0.5}SiB_2$ is a type-II superconductor. The lower critical field $H_{c1}(T)$ was determined from the magnetic field versus magnetization (*M-H*) curves at various temperatures. As shown in Fig. 1, $H_{c1}(T)$ was defined as a magnetic field at which the diamagnetic magnetization deviates from a linear relation with the magnetic field. $H_{c1}(T)$ is shown in Fig. 1(b) as a function of $(T/T_c)^2$, $H_{c1}(T)$ was fitted by the relation $H_{c1}(T) = H_{c1}(0)[1-(T/T_c)^2]$, following the Ginzburg Landau (GL) theory, where $H_{c1}(0)$ is $H_{c1}(T)$ at 0 K. H_{c1} was determined to be about 121 Oe. The penetration depth λ was calculated to be approximately 369 nm from the relationship between H_{c1} and λ, $\mu_0 H_{c1}(T) \sim \Phi_0/\pi\lambda^2$, where μ_0 and Φ_0 are the magnetic permeability of the vacuum quantum flux, respectively.

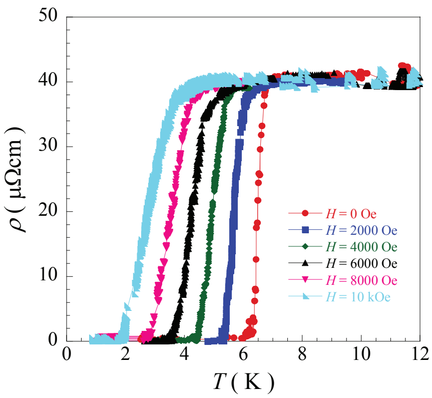

Fig. 2. (Color online) Temperature dependence of electrical resistivity (measuring current $I = 1$ mA) of $W_{4.5}Ta_{0.5}Si_{2.1}B_2$ under various magnetic fields.

Figure 2 shows the temperature dependence of resistivity under various magnetic fields up to 10 kOe. T_c^{onset} (onset transition temperature) and T_c^{zero} (zero-resistivity temperature) decrease with increasing applied magnetic field. The transition width remains almost unchanged up to high magnetic fields. T_c is determined at a midpoint of transition, and transition width is taken as the temperature interval between 10 and 90% of transition.

Figure 3 shows the temperature dependence of critical field $H_{c2}(T)$ (H-T phase diagram) for $W_{4.5}Ta_{0.5}SiB_2$. H_{c2} was determined by electrical resistivities under various magnetic fields. On the basis of the relation $H_{c2}(T) = H_{c2}(0)[1-(T/T_c)^{3/2}]^{3/2}$ for the type-II superconductor[5], $H_{c2}(0)$ is found to be about 15 kOe. This $H_{c2}(0)$ value is clearly below the Pauli limit of H_p = 18.3 (kOe/K) $T_c \approx$ 120 kOe (assuming a weak-coupling limit). From these data, $\xi(0)$ is calculated to be ~15 nm using the relation $H_{c2}(0) = \Phi_0/\pi\xi^2$, where Φ_0 is the flux quantum. The GL parameter κ_{GL} is estimated to be 24.7 using the relation $\kappa_{GL} = \lambda/\xi$. Moreover, the thermodynamic critical field $H_c(0)$ was calculated to be approximately 4220 Oe on the basis of the relation $H_{c1}H_{c2} = H_c^2$.

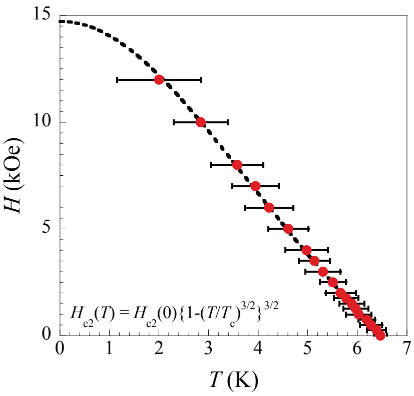

Fig. 3. (Color online) *H-T* phase diagram deduced from electrical resistivity of $W_{4.5}Ta_{0.5}Si_{2.1}B_2$ under various magnetic fields. The curve shows a fit to the local-pairing theory.

Figure 4 shows the temperature dependence of electronic specific heat C_{el} of $W_{4.5}Ta_{0.5}SiB_2$ at H = 0 Oe. The inset figure of Fig. 4 shows the normal-state specific heat obtained by applying a magnetic field H = 20 kOe ≥ H_{c2}. At H = 0 Oe, specific heat data shows the bulk superconducting feature, as indicated by the clear jump of C_{el} at around T_c. T_c of 6.5 K is in good agreement with the value obtained from other measurements. At H = 20 kOe, the specific heat jump, is completely suppressed, resulting in the disappearance of superconductivity in $W_{5-x}Ta_xSiB_2$. As shown in Fig. 4, in order to take into account the normal-state C_{ph} (lattice part), we fitted the data obtained at H = 20 kOe by a polynomial, $C_P/T = \gamma_N + \beta T^2$, with the Sommerfeld coefficient of the normal-state specific heat γ_N and the coefficient of the phononic contribution β. β is related to the Debye temperature θ_D via $\beta = (12/5)N\pi R\theta_D^{-3}$, where R = 8.314 J/(mol K) denotes the gas constant and N = 8 is the number of atoms per formula unit for $(W, Ta)_5SiB_2$.

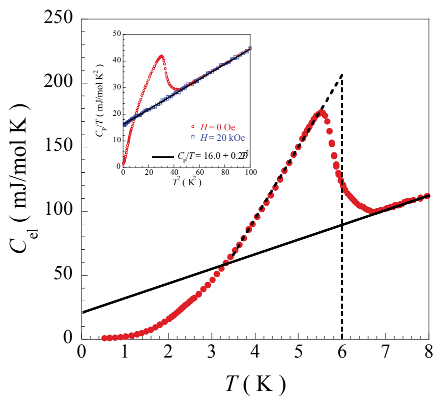

Fig. 4. (Color online) Temperature dependence of electrical specific heat of $W_{4.5}Ta_{0.5}Si_{2.1}B_2$. The inset shows the specific heat at H = 0 and 20 kOe. The solid line in the inset shows a fit to the experimental data using the Debye formula.

The specific-heat measurements were carried out on a sample, which was also characterized by powder X-ray diffraction analysis. The sample contained two phases: $W_{4.5}Ta_{0.5}SiB_2$ (the main phase) and WSi_2 (the minor phase). The contribution of the sample to the measured specific heat C_P can be determined using C_P(μJ/(K g))=yC_P

$^{W4.5Ta0.5SiB2}$+(1-y) C_P^{WSi2}, where y and 1-y are the weight fractions of $W_{4.5}Ta_{0.5}SiB_2$ and WSi_2. We actually measured the specific heat data of WSi_2, and the γ_N and β of WSi_2 were determined to be 1.36(1) and 0.03(1) mJ/(mol K^4), respectively. We carefully subtracted the contribution of WSi_2 to total heat capacity. From this fit, we deduced γ_N = 16.0 mJ/(mol K^2) and $\beta = 0.29$ mJ/(mol K^4). We calculated the corresponding Debye temperature θ_D to be approximately 380 K. In the superconducting state, the electronic specific heat C_{el} of $W_{4.5}Ta_{0.5}SiB_2$ can be obtained after subtracting the phononic contribution from the total specific heat: $C_{el} = C_P^{W4.5Ta0.5SiB2}$-$C_{ph}$. The C_{ph} data was obtained from the fitting result. The normalize specific heat jump $\Delta C/\gamma_N T_c$ is calculated to be about 1.22 using γ_N = 16 mJ/(mol K^2) and T_c = 6 K considering entropy balance.

Finally we discuss the enhancement of T_c between W_5SiB_2 and $W_{4.5}Ta_{0.5}SiB_2$. The $\Delta C/\gamma_N T_c$ value of $W_{4.5}Ta_{0.5}SiB_2$ and W_5SiB_2 ($\Delta C/\gamma_N T_c$ = 1.49) is much closer to that of predicted $\Delta C/\gamma_N T_c$ =1.43 in BCS theory. Consequently, superconducting state in$W_{4.5}Ta_{0.5}SiB_2$ also exists in weak coupling regime and can be described in a conventional BCS framework. In the weak-coupling BCS theory, T_c is given by the equation $k_B T_c = \hbar\omega_{ph}\exp\{-1/VN(E_F)\}$, where ω_{ph} denotes the frequency of the relevant phonons, which is proportional to θ_D, $N(E_F)$ is the density state of Fermi level, and V and V' are the electron–phonon coupling strength for W_5SiB_2 and $W_{4.5}Ta_{0.5}SiB_2$, respectively [6]. The different values of T_c possess information on the difference in the density of states at the Fermi level $N(E_F)$ for the two compounds given by $V'/V = \{N(E_F)\log(T_c/\omega_{ph})\}/\{N'(E_F)\log(T_c'/\omega_{ph}')\}$, where we assume $N(E_F)/N'(E_F) = \gamma_N/\gamma_N'$. This leads to $V'/V = \sim 0.86$ using T_c = 5.8 K, θ_D = 470 K, and γ_N = 12.8 mJ/(mol K^2) for W_5SiB_2 and T_c' = 6.5 K, θ_D' = 380 K, and γ_N' = 16 mJ/(mol K^2) for $W_{4.5}Ta_{0.5}SiB_2$. The T_c value of $W_{4.5}Ta_{0.5}SiB_2$ would have been much higher if the electron–phonon coupling strength were not reduced by approximately 14%. θ_D value of is higher than that of W_5SiB_2. These facts suggest that the phononic contribution of enhancement of T_c is small. In previous study of band structure calculation suggests that the W d-orbital mainly contributed to density of state near Fermi level [7]. γ_N value $W_{4.5}Ta_{0.5}SiB_2$ is higher than that of W_5SiB_2. This fact indicates that $N(E_F)$ of W_5SiB_2 is enhancement by substituting Ta for W. Moreover, an enhancement of $N(E_F)$ mainly contribute to enhancement of T_c in (W, Ta)$_5$SiB$_2$ system.

4. Summary

We investigated the superconducting state of $W_{5-x}Ta_xSiB_2$ with T_2 phase structure. We revealed its superconducting and normal-state properties in detail by means of magnetic susceptibility, electrical resistivity, and magnetization measurements. The magnetization as a function of magnetic field showed a type-II superconducting behavior, and $H_{c1}(0)$ and λ were estimated to be 121 Oe and 369 nm, respectively. The $H_{c2}(0)$ values were estimated to be 15 kOe from the temperature dependence of resistivity under several magnetic fields. The $\xi(0)$ value was calculated to be approximately 15 nm from $H_{c2}(0)$. From the $\xi(0)$ and $\lambda(0)$ values, κ was estimated to be 24.7. Specific heat measurements indicated that $W_{5-x}Ta_xSiB_2$ can be described as a conventional BCS superconductor in the week-coupling regime: it has typical thermal properties: $\Delta C/\gamma_N T_c$ = 1.22. We concluded that the enhancement of T_c in $W_{5-x}Ta_xSiB_2$ system induced the increasing density of state near E_F by partially substituting Ta for W.

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